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(54) NEGATIVE ELECTRODE OF LITHIUM ION SECONDARY BATTERY AND LITHIUM ION SECONDARY BATTERY

(57)Abstract:

PROBLEM TO BE SOLVED: To provide a negative electrode of a lithium ion secondary battery whose life can be prolonged with keeping a high initial coulomb efficiency specific to cylindrical graphite carbon fiber, and to provide a lithium ion secondary battery using the same.

SOLUTION: The negative electrode of a lithium ion secondary battery in relation to the invention mainly comprises a composite body wherein cylindrical graphite carbon fiber is mixed with scale-like graphite powder. A ratio of the scale-like graphite powder in the mixed composite is from 20 to 50 weight percentages, granular diameter thereof is from 0.1 to 30 μm , and granular diameter of the cylindrical graphite carbon fiber is from 5 to 50 μm . In the negative electrode in relation to the invention, inner stress accompanied by compression and shrink caused by repetitive use can be dispersed owing to the existence of both cylindrical graphite carbon fiber and the scale-like graphite powder so that the negative electrode is not deformed. Therefore, the lithium ion battery including the negative electrode and a positive electrode formed of an appropriate material can express excellent cyclic properties.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention]This invention relates to the rechargeable lithium-ion battery itself [concerned], concerning the negative electrode of a rechargeable lithium-ion battery.

[0002]

[Description of the Prior Art]The conventional negative electrode for rechargeable lithium-ion batteries makes the end of carbon powder adhere on a metal electrode using binders, such as polyvinylidene fluoride (PVdF), and is created. Depending for the characteristic on material structures, such as particle diameter of a carbon material, shape, a degree of crystallinity, and its stacking tendency, greatly, when using these carbon materials as an electrode material is known, and it is known that the structure of a carbon material will take various gestalten according to manufacturing conditions, such as a starting material and calcination.

[0003]If the performance of the cell to constitute is taken into consideration, charge-and-discharge capacity density, initial coulomb efficiency, a high rate discharging characteristic, and a cycle characteristic are important as the characteristic of the negative electrode using a carbon material. Although the electrode characteristic of the carbon material of former versatility has been examined, the material in which the characteristic outstanding in these [all] is shown is not found.

[0004]Generally, the graphite material with large particle diameter with a high degree of crystallinity has a tendency which shows high initial coulomb efficiency. The cylindrical graphitized carbon fiber which calcinates carbon fiber in the elevated temperature of about 3000 degrees, and is compounded especially has the small irreversible capacity at the time of first time charge and discharge, it excels in a high rate discharging characteristic, and an initial characteristic suitable as a negative electrode of a rechargeable lithium-ion battery is given.

[0005]

[Problem(s) to be Solved by the Invention]However, since the negative electrode using the above cylindrical graphitized carbon fibers did not have the one-dimensional direction of expansion and contraction of the active material accompanying charge and discharge, it was easy to localize the stress inside an electrode, and the technical problem that it was easy to produce a problem from a viewpoint of reinforcement occurred.

[0006]On the other hand, the negative electrode using scale-like black lead has the small capacity lowering accompanying advance of a charging and discharging cycle, and the outstanding cycle characteristic is shown. However, in order that the crystal axis of the active material inside an electrode may carry out orientation of the negative electrode using this material to a specific direction, expansion and the contracting direction of an electrode turn into one way at the time of charge and discharge. For this reason, there was a problem that the diffusion length of a lithium ion with low initial coulomb efficiency became large, and a high rate discharging characteristic was bad since the specific surface area of material which modification of an electrode and a cell tends to generate is large when expansion and a shrinkage amount cell-ize greatly.

[0007]The place which this invention was made in light of the above-mentioned circumstances, and is made into the purpose, Although high initial coulomb efficiency peculiar to a cylindrical graphitized carbon fiber, small expansion and shrinkage amount, and the outstanding high rate discharging characteristic are maintained, it is in providing the negative electrode of the rechargeable lithium-ion battery in which the reinforcement in the use is possible, and a rechargeable lithium-ion battery.

[0008]

[Means for Solving the Problem]This invention took the following means, in order to solve the above-mentioned technical problem. That is, a negative electrode of the rechargeable lithium-ion battery according to claim 1 makes main components mixed composite of a cylindrical graphitized carbon fiber and scaly graphite powder.

[0009]According to this, stress inside the negative electrode concerned by which it is generated by repeating charge and discharge is distributed by putting expansion and contraction of the direction of two dimensions in a cylindrical graphitized carbon fiber, and expansion and contraction of the direction of one dimension by scaly graphite powder together. That is, the negative electrode concerned obtains an operation that whose it changes easily etc. it is lost. According to this negative electrode, invention-in-this-application persons checked that improvement in other characteristics concerning a rechargeable lithium-ion battery, such as first time service capacity density, could also be expected.

[0010]As said scaly graphite powder in a negative electrode of a rechargeable lithium-ion battery mentioned above, A rate that it occupies in said mixed composite is 20 to 50 % of the weight (claim 2), When fulfilling a monograph affair of being [the particle diameter is 0.1-30 micrometers, and / particle diameter of said cylindrical graphitized carbon fiber / 5-50 micrometers

(claim 3)] **, it checked that it was also possible to employ efficiently an operation mentioned above more effectively. A negative electrode of the rechargeable lithium-ion battery according to claim 4 is the invention which caught the above-mentioned matter from an operating surface.

[0011]With an anode which consists the above-mentioned negative electrode of a metal chalcogen ghost, a metallic oxide, a lithium multiple oxide, a conjugated system high molecular compound that has conductivity, etc. If constituted as a rechargeable lithium-ion battery (claim 5), since modification of a negative electrode in repetition use does not arise, in the cell concerned, improvement in service capacity density and a cycle characteristic, etc. can be expected.

[0012]

[Embodiment of the Invention]Below, an embodiment of the invention is described. In the nonaqueous electrolyte type rechargeable lithium-ion battery with which the rechargeable lithium-ion battery concerning this invention uses an anode, a negative electrode, an electrolysis solution, a separator, etc. as the main components, A negative electrode consists two sorts of different carbon materials, i.e., scaly graphite powder, and a cylindrical graphitized carbon fiber of mixing and a composite-sized thing. These each component is explained below.

[0013]First, as positive active material, a thing which is illustrated next can be chosen so that it may usually be used.

** TiS₂, MoS₃, NbSe₃, FeS, VS₂, the metal chalcogen ghost that has the layer structure of VSe₂ grade.

** The metallic oxide of CoO₂, Cr₃O₅, TiO₂, CuO, V₃O₆, MoO₃, V₂O₅, and Mn₂O₃.

** The multiple oxide which replaced a part of LiCoO₂, LiNiO₂, LiMn₂O₄, Co in each, nickel, or Mn with other elements, for example, Co, Mn, Fe, nickel, etc. further.

** The conjugated system polymers combination substance which has conductivity, such as polyacetylene, poly aniline, poly para-phenylene, a polythiophene, and polypyrrole.

[0014]As an electrolysis solution, for example Propylene carbonate, ethylene carbonate, Gamma-butyrolactone, a tetrahydrofuran, 2-methyltetrahydrofuran, Dioxolane, 4-methyl dioxolane, sulfolane, 1, 2-dimethoxyethane, Dimethyl carbonate, diethyl carbonate, dimethyl sulfoxide, Acetonitrile, N,N-dimethylformamide, a diethylene glycol, To aprotic solvents, such as wood ether, or the mixed solvent of two or more sorts of these solvents. LiBF₄, LiClO₄, LiAsF₆, LiSbF₆, LiAlO₄, LiAlCl₄, LiPF₆, LiN (C_xF(2x+1) SO₂) (CyF(2y+1) SO₂) (x and y are natural numbers), LiCl, LiI, etc. can be used as an electrolyte. These may be used, where it could use independently and two or more sorts are mixed. The solid electrolyte which distributed the above-mentioned electrolyte can be used instead of an electrolysis solution.

[0015]The porous membrane of the polyolefin system of nonwoven fabrics, such as porosity polypropylene by which normal use is carried out, etc. can be used for a separator.

[0016]next, the negative electrode concerning this invention -- the carbon material, i.e., the scaly graphite powder, and cylindrical graphitized carbon fiber of two sorts of different shape -- mixing -- it composite-sizes and creates. By the way, the initial characteristics and cycle characteristics (measured as a "degradation rate".) of a negative electrode, such as service capacity density, irreversible capacity density, and coulomb efficiency As for this unit, "%/cycle" is greatly influenced by the kind of carbon material which constitutes a negative electrode, shape, an addition (mixing ratio), particle diameter, and a composite-sized method. Here, it can express the service capacity density of the first stage, irreversible capacity density, and coulomb efficiency as what fills the following relation.

[0017]

Irreversible capacity density = initial coulomb efficiency [of first-time-charge capacity density-first time service capacity density] = (first time service capacity density / first-time-charge capacity density) x100=(first time service capacity density/(first time service capacity density + irreversible capacity density)) x100 [0018]Therefore, coulomb efficiency becomes large, so that irreversible capacity density is low. A degradation rate serves as a decision criterion of a cycle characteristic, and shows the difference of the service capacity density at the time of the first time, and the service capacity density at the time of the number of times of a constant cycle by the percentage per 1 cycle (%). Therefore, a degradation rate is so good that it is small.

[0019]By using together the cylindrical graphitized carbon fiber which serves as material of the above-mentioned negative electrode in this invention, and scale-like graphite powder, and controlling each addition and particle diameter of these, The direction of expansion and contraction of the material at the time of charge and discharge and its size are controlled, the modification of an electrode and the concentration of stress accompanying a charging and discharging cycle are made as small as possible, and improvement in a cycle characteristic is attained, without spoiling high peculiar to cylindrical graphitized carbon fiber first stage coulomb efficiency, and the outstanding high rate discharging characteristic.

[0020]It is as follows if the above-mentioned situation is explained more concretely. First, that to which the term of "cylindrical shape" here thru/or the "shape of a scale" points is a comparatively macroscopic gestalt which respectively very small microcrystals gather and is formed. Drawing 1 and drawing 2 express typically the state where the gestalt about these both carried out cross sectional view, and the numerals [in / for the section of "cylindrical shape" graphitized carbon fiber / in the numerals 11 in drawing 1 / drawing 2] 12 show the section of "scale-like" graphite powder, respectively. the above [in / in a difference of these gestalten / both] -- the difference of the orientation of very small microcrystal is also expressed. That is, the cylindrical graphitized carbon fiber shown in drawing 1 has the composition that the graphite-crystals layer was allotted circularly and in multilayer on the basis of the center of the circular section. In other words, it can be called the state where the crystal orientation of very small microcrystal is carrying out orientation in parallel with a cylinder medial axis. On the other hand, scaly graphite powder is in the state where the crystal orientation of very small microcrystal was allotted almost uniformly within the 1 scale. Namely, it is in the state where graphite-crystals layers overlap in parallel.

[0021]By the way, generally the charge and discharge to a graphite material are a phenomenon in which a lithium ion goes in

and out between the graphite-crystals layers which constitute the graphite material concerned. In that case, although this is called what is called an intercalation reaction, as the distance between the graphite-crystals layers 13 is shown in [drawing 3](#), it will change according to receipts and payments of the lithium ion 14, i.e., charge and discharge. Although it is needless to say, the state where the lithium ion 14 enters between the graphite-crystals layers 13, and the distance between the graphite-crystals layers 13 becomes large is the reverse state at the discharge time at the time of charge.

[0022]When based on these matters, it turns out that change of the distance between the above-mentioned graphite-crystals layers shows a mode which is different with a cylindrical graphitized carbon fiber and scaly graphite powder, respectively. That is, in the former, as shown in [drawing 1](#), it will appear as a mode which is expanded and contracted in radiation to the center of the section which becomes circular, and as shown in [drawing 2](#) (inside of a figure), in one scaly graphite powder, it will appear in a sliding direction as a mode of expanding and contracting. That is, it can say that it is two-dimensional in expansion and contraction of a cylindrical graphitized carbon fiber, and it of scaly graphite powder is one-dimensional.

[0023]And in this invention, by combining these cylindrical graphitized carbon fibers and scaly graphite powder, expansion and contraction of a result and the direction of two dimensions, and expansion and contraction of the direction of one dimension will be combined, the stress inside a negative electrode will be distributed, and the above-mentioned purpose can be attained.

[0024]In order to attain the above-mentioned purpose better, it is preferred to fulfill the conditions explained below. When the addition of scaly graphite powder shows this first as the desirable mixing ratio of the two above-mentioned sorts of carbon materials, 20 to 50 % of the weight is desirable. It is because initial coulomb efficiency will fall if a cycle characteristic gets worse at less than 20 % of the weight in an addition and an addition exceeds 50 % of the weight. This is considered because scaly graphite powder is playing a role of a conducting material, and a role of the stress distribution in an electrode in a compound carbonizing matter negative electrode. An addition here is weight [when what will be in the state where the binder (after-mentioned) for constituting a negative electrode was removed is made into entire weight] %.

[0025]Next, about the particle diameter of two sorts of carbon materials used for a negative electrode, it is preferred to fulfill the following conditions. That is, as mean particle diameter of scaly graphite powder, 0.1-30 micrometers is good in it being 0.1-20 micrometers still more preferably. Because, it is for initial coulomb efficiency to fall, if particle diameter is set to less than 0.1 micrometer, and for service capacity density to fall, if particle diameter exceeds 30 micrometers. When calling it particle diameter in this embodiment, the above-mentioned mean particle diameter shall be meant and this mean particle diameter shall mean further the "average" particle diameter as the whole sample measured by laser diffractometry. This situation is the same also in the example described below.

[0026]On the other hand, as particle diameter of a cylindrical graphitized carbon fiber, 5-50 micrometers is desirable. The reason is the same as that of the aforementioned case almost, if particle diameter is set to less than 5 micrometers, initial coulomb efficiency will fall substantially, and particle diameter is because service capacity density falls in not less than 50 micrometers and it stops showing a practical charging and discharging characteristic.

[0027]The following situations are pointed out about the particle diameter of the above-mentioned scaly graphite powder and a cylindrical graphitized carbon fiber. That is, although it is determined for reasons of decline in coulomb efficiency that each of both lower limit (0.1 micrometer and 5 micrometers) mentioned above, this has the following backgrounds. That is, since the cause of a manifestation of irreversible capacity is considered to originate in the decomposition reaction of the electrolysis solution in the carbon particle surface, it is that specific surface area decreases, irreversible capacity decreases while particle diameter increases, namely, it is thought that coulomb efficiency falls. The thing as which it requests that such a situation has a certain amount of [particle diameter] size and which the suitable minimum particle diameter exists if it puts in another way is meant, and both the above-mentioned lower limits are decided to fulfill this condition. Since the sedimentation velocity of the particles in a slurry (after-mentioned) will become large if particle diameter besides the reason of the fall of above-mentioned service capacity density becomes not much large about both the above-mentioned upper limit (30 micrometers and 50 micrometers), The situation that it is difficult to obtain a slurry with the characteristic suitable for production by being stabilized is also taken into consideration.

[0028]Next, the mixed composite-ized method of scaly graphite powder and a cylindrical graphitized carbon fiber is explained. The various molding methods can be used for this composite-ization, and after adding a binder to both mixture and mixing, the various molding methods, such as extrusion, ejection, and compression, are applicable. As a method of incidentally forming both mixture before resulting in molding (i.e., before resulting in composite-ization), a liquid phase mixed method, a solid phase mixed method, etc. which use a solvent can be used. As a mixing method of a liquid phase process, the various mixed methods of a homomixer, a ball mill, an ultrasonic dispersion machine, a jet mill mixer, etc. are applicable. Polyvinylidene fluoride, polyethylene powder, various rubber system materials other than polytetrafluoroethylene, etc. can be used for a binder, for example.

[0029]The negative electrode composite-ized [above] can be used with various gestalten according to the shape and the use of a cell. For example, the shape of a disk, tabular, the shape of a film, film state, a sheet shaped, etc. can be considered. According to shape, a use, and a gestalt, it is selectable suitably also in the thickness of a negative electrode.

[0030]The negative electrode for rechargeable lithium-ion batteries of this invention manufactured as mentioned above can be used as a rechargeable lithium-ion battery by combining with positive active material and nonaqueous electrolyte suitably.

[0031]

[Example]Below, the example based on this invention which invention-in-this-application persons checked is described more concretely. Drawing of longitudinal section of the coin type nonaqueous electrolyte secondary battery for evaluating the negative electrode of this invention is shown in [drawing 4](#). In a figure, the cell case 21 processes and forms the stainless

steel plate of organic electrolysis-proof acidity or alkalinity. The lid 22 constituted with stainless steel as well as that material is covered and installed in this cell case 21. As for the anode 23, metal lithium was being stuck to the charge collector made from stainless steel by pressure. And as shown in a figure, the carbon negative electrode 24 used as the characterizing portion of this invention is formed so that this anode 23 may be countered in directly under [of said lid 22], and the upper part of said anode 23. Between these carbon negative electrodes 24 and the anode 23, it has the separator 26 of the filter paper 25 made from glass wool, and the fine porous film made from polypropylene. It has the gasket 27 made from polypropylene so that the side of these composition may be covered (to circumference shape [Facing from the upper surface.]).

[0032]The electrolysis solution dissolved tetrafluoride lithium borate in the mixed solvent of carbon ethylene:carbon dimethyl =1:2 by the concentration of 1 mol/l. as a solute, and was used for it. The size in the cell of this example incidentally constituted in this way is 20 mm in diameter, and 1.6 mm in height.

[0033]As a carbon material used for the carbon negative electrode 24, product mesophase pitch system graphitized carbon fiber Mild (the mean particle diameter of 18 micrometers, fiber diameter of 9 micrometers) made by PETOKA of marketing as a cylindrical graphitized carbon fiber -- commercial Lonza scale-like artificial-graphite powder (mean particle diameter of 6 micrometers) was used as a different carbon material from this, respectively. The carbon negative electrode 24 was created as follows by these. Namely, what mixed both the materials mentioned above as a ratio of 80:20, 50:50, and 20:80 by the weight ratio is prepared, respectively, Polyvinylidene fluoride powder was set to carbon material mixture:binder =100:10 (weight ratio) as a binder, and to these mixtures, a proper quantity of N-methyl pyrrolidone was added as mixing and also a solvent, and it mixed into them, and slurred into them. Next, using the doctor blade method, it dried on one side of copper foil (20 micrometers in thickness) after applying to homogeneity, compression molding (composite-izing) of the obtained slurry was carried out to it, and the carbon negative electrode 24 16 mm in diameter which it pierces circularly and is net 100 micrometers in thickness was created for this. The structure gestalt of carbon negative electrode 24 inside formed in this way is shown as a copy figure which expressed the SEM image realistically, as shown in [drawing 5](#). This figure shows having the structure where the scaly graphite powder 12 was arranged on the circumference of the cylindrical graphitized carbon fiber 11, and both were composite-ized. This was made the carbon negative electrode 24 created as mentioned above and the anode 23 with the testing cell by attaching a lead, respectively.

[0034]In this example, the above-mentioned testing cell was used and the constant-current charge test was done as charge and discharge current density 100 mAh/g-C under with the charge final voltage 1.5V and a discharge final voltage of 5 mV conditions. It examined on the same conditions as the above also with the case where a cylindrical graphitized carbon fiber and a scale-like artificial graphite use the carbon negative electrode 24 which it comes to constitute independent more, respectively, for comparison. About the particle diameter of scaly graphite powder, by two or more sorts (0.1, 5, 10, 15 and 20, and 30 micrometers) in within the limits which is 0.1-30 micrometers, it created the testing cell and examined similarly. Below, suppose that it explains with reference to these, those results being shown in [drawing 8](#), [drawing 9](#), and [drawing 10](#) from Table 1 and [drawing 6](#).

[0035]First, addition of scaly graphite powder explains the influence which it has on the characteristic of a rechargeable lithium-ion battery. Table 1 shows this collectively.

[0036]

[Table 1]

電池番号	鱗片状黒鉛 粉末の添加量	初期特性			劣化率
		放電容量密度	不可逆容量密度	クーロン効率	
	[wt%]	[Ah/kg]	[Ah/kg]	[%]	[%/cycle]
1	0	280	23	92.3	1.752
2	20	300	45	87.0	0.100
3	50	327	57	85.1	0.069
4	80	360	109	76.8	0.264
5	100	330	130	71.7	0.177

[0037]It is needless to say and the state where all the positive active material is scaly graphite powder is suggested, saying "100 % of the weight of additions". [in / here / Table 1]

[0038]Service capacity density is 280 by comparing with the other thing that whose addition of scaly graphite powder is 0 % of the weight according to this table 1. [Ah/kg] ** et al. [300] [Ah/kg] It becomes the above and, in improvement in the characteristic, a degradation rate is 1.752 again. [%/cycle] ** et al. [0.264] [%/cycle] It becomes the following and the sharp fall can be grasped. However, when the addition of scaly graphite powder is made into 80 % of the weight or more, it turns out that irreversible capacity density will rise, and coulomb efficiency will also fall compared with the case where the addition considers it as 20 thru/or 50 % of the weight, and a degradation rate also gets worse.

[0039][Drawing 6](#) - [drawing 8](#) show the above-mentioned situation as a graph. [Drawing 6](#) is the graph which showed each of the difference (cell numbers 1-5 in Table 1) of the addition of scaly graphite powder by change of the occasional service capacity density about the testing cell appropriated for the repetition use first. When this is seen, that the difference of an action with the other thing has appeared very clearly turns out to be 0 % of the weight of additions. [Drawing 7](#) is the graph which showed change of the irreversible capacity density to the addition of scaly graphite powder.

[Drawing 8](#) is the graph which similarly showed change of the coulomb efficiency over the addition of scaly graphite powder. The dotted line shown in [drawing 7](#) and [drawing 8](#) expresses that it is preferred that irreversible capacity density is below the dotted line concerned with the arrow shown by aligning with a dotted line in [drawing 7](#).

Also in drawing 8, it expresses that it is preferred similarly that coulomb efficiency is more than the dotted line concerned.

[0040]It is clear from the above thing that the addition's of scaly graphite powder 20 to 50 % of the weight is the most desirable conditions. That is, service capacity density and coulomb efficiency are large, a degradation rate is small, namely, by fulfilling this condition shows that the rechargeable lithium-ion battery which was excellent in the cycle characteristic can be provided.

[0041]next, the mean particle diameter of scaly graphite powder does to the characteristic of a rechargeable lithium-ion battery -- influence ***** explanation is given. Drawing 9 is a graph which drawing 10 is the same again and shows the relation between the mean particle diameter of scaly graphite powder, and irreversible capacity density for the relation between the mean particle diameter of scaly graphite powder, and first time service capacity density, respectively. When particle diameter is set to not less than 20 micrometers so that clearly from drawing 9, it turns out that first time service capacity density falls. It turns out that first time service capacity density especially sinks below 200 Ah/kg with the particle diameter of not less than 30 micrometers, and it gets worse remarkably. Although the fall of irreversible capacity density can be read in drawing 10 with the increase in particle diameter, it combines also with the consideration in drawing 9, and the mean particle diameter of scaly graphite powder can be called after all 0.1-30 micrometers and more desirable thing which has desirable 0.1-20 micrometers.

[0042]

[Effect of the Invention]As explained above, the negative electrode of the rechargeable lithium-ion battery according to claim 1, Since mixed composite of a cylindrical graphitized carbon fiber and scaly graphite powder is made into main components, modification resulting from the internal stress by repetition use of the negative electrode [concerned] concerned can be prevented by mixing that from which compression and contraction take place in the direction of one dimension thru/or the direction of two dimensions. According to this negative electrode, the improvement in first time service capacity density etc. is also attained.

[0043]As said scaly graphite powder in the negative electrode of the rechargeable lithium-ion battery mentioned above, The rate that it occupies in said mixed composite is 20 to 50 % of the weight (claim 2), If the monograph affair of being [the particle diameter is 0.1-30 micrometers, and / the particle diameter of said cylindrical graphitized carbon fiber / 5-50 micrometers (claim 3)] ** is fulfilled, the effect mentioned above can be enjoyed more certainly. It is clear that the negative electrode's of the rechargeable lithium-ion battery according to claim 4 each above-mentioned effect is enjoyable.

[0044]With in addition, the anode which consists the above-mentioned negative electrode of a metal chalcogen ghost, a metallic oxide, a lithium multiple oxide, a conjugated system high molecular compound that has conductivity, etc. If constituted as a rechargeable lithium-ion battery (claim 5), since modification of the negative electrode in repetition use does not arise, in the cell concerned, improvement in service capacity density and a cycle characteristic, etc. can be attained.

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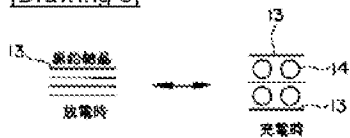
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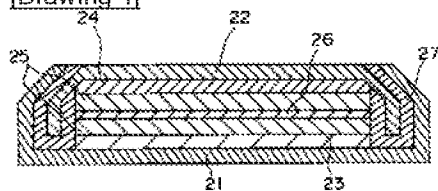
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DRAWINGS

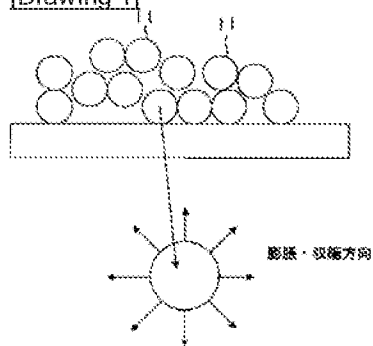
[Drawing 3]



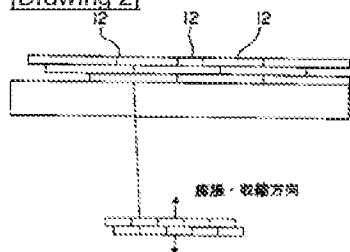
[Drawing 4]



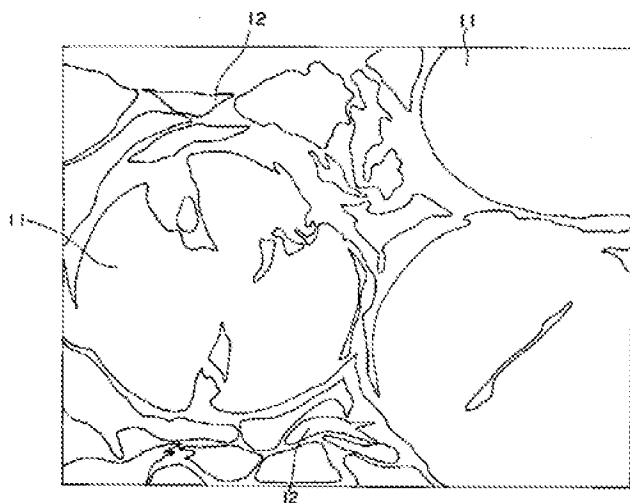
[Drawing 1]



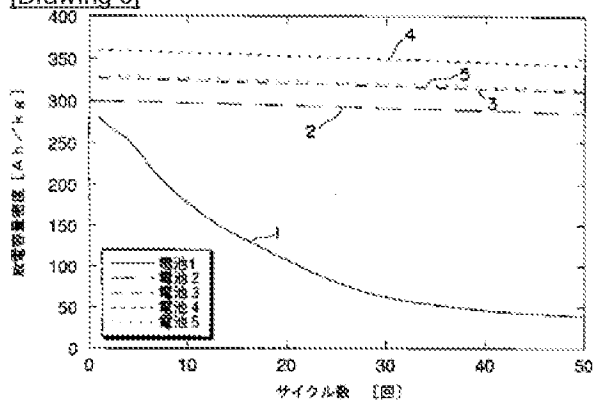
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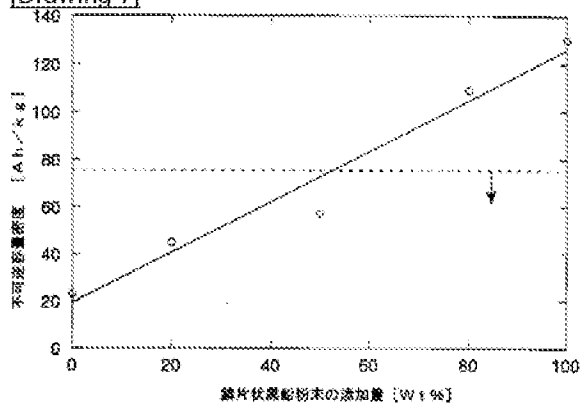
[Drawing 5]



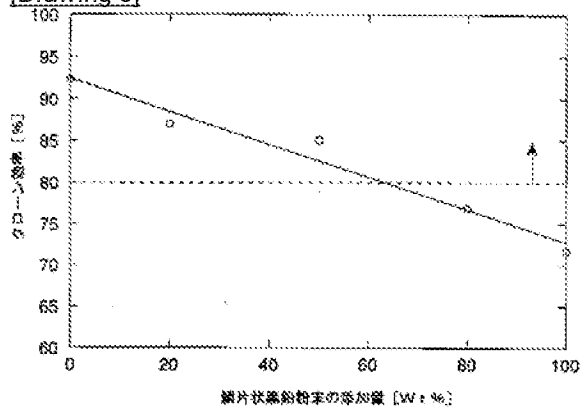
[Drawing 6]



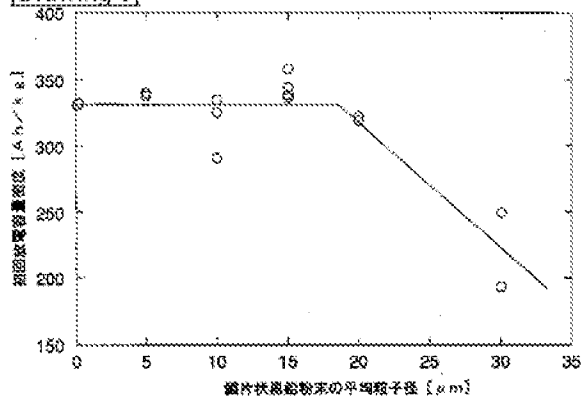
[Drawing 7]



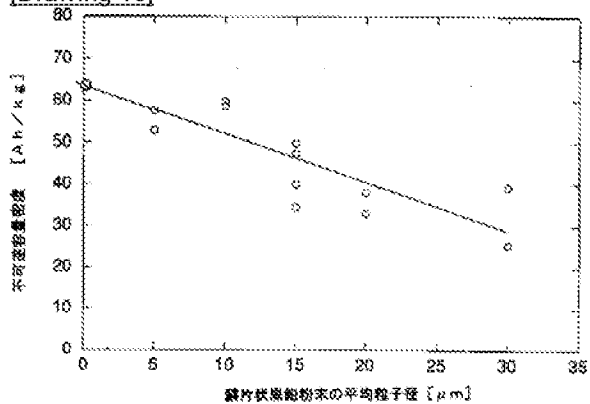
[Drawing 8]



[Drawing 9]



[Drawing 10]



[Translation done.]